Angular Dependence of the Superconducting Transition Temperature in Ferromagnet-Superconductor-Ferromagnet Trilayers

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The superconducting transition temperature, T_c, of a ferromagnet (F) - superconductor (S) - ferromagnet trilayer depends on the mutual orientation of the magnetic moments of the F layers. This effect has been previously observed in F/S/F systems as a T_c difference between parallel and antiparallel configurations of the F layers. Here we report measurements of T_c in CuNi/Nb/CuNi trilayers as a function of the angle between the magnetic moments of the CuNi ferromagnets. The observed angular dependence of T_c is in qualitative agreement with a F/S proximity theory that accounts for the odd triplet component of the condensate predicted to arise for non-collinear orientation of the magnetic moments of the F layers.

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Electronic properties of a superconducting (S) film can be significantly altered by direct contact with a metallic ferromagnet (F) [1, 2]. Penetration of the superconducting condensate into a ferromagnet results in the breaking of singlet Cooper pairs by the exchange field and leads to a decrease of the superconducting transition temperature (T_c). Recent discoveries of quantum interference effects in F/S heterostructures have shown that the F/S proximity effect is more interesting and complex than just the reduction of T_c [3–9]. Examples of non-trivial F/S proximity phenomena include (i) oscillations of T_c with the F layer thickness (d_f) in F/S multilayers [3, 4], (ii) oscillations of the critical current in S/F/S Josephson junctions with d_f [5] and (iii) manipulation of T_c in F/S/F [6, 7] and F/N/F/S [8, 9] structures (N denotes a nonmagnetic metal) by switching the magnetic state of the F layers.

The superconducting condensate of a F/S system is predicted to consist of a singlet component and a triplet component with unusual symmetry [10–16]. The triplet part of the condensate function is expected to be s-wave (i.e. even in the momentum variable). Therefore, by the Pauli exclusion principle, it must be odd in time. The s-wave character of the condensate makes it robust against impurity scattering, and thus the odd triplet pairing should survive even in dirty F/S systems. If the magnetization of the ferromagnet is not spatially uniform, the triplet part of the condensate has three non-zero components corresponding to three projections, $S_z = (0, \pm 1)$ of the Cooper pair spin. The $S_z = \pm 1$ components are immune to pair breaking by the exchange field, and thus can penetrate deep into the ferromagnet [17]. Recent measurements of the critical current in Josephson junctions with noncollinear magnetic barriers support the existence of this long-range odd triplet superconductivity [18].

The simplest model system for studies of the effect of spatially non-uniform magnetization on superconductivity is a F/S/F trilayer with non-collinear (neither parallel nor antiparallel) orientation of the magnetic moments of the F layers [7, 13]. However, experimental tests of the F/S proximity effect in this system have been limited to the collinear geometry [6, 19–22]. In this Letter, we describe experimental and theoretical studies of T_c in CuNi/ Nb/ CuNi trilayers as a function of the angle between the CuNi layer magnetizations. We compare our experimental results to theories of the F/S proximity effect that take into account the odd triplet component of the condensate, and find that the clean limit of the proximity effect theory gives good qualitative description of the data.

The multilayer samples for our experiments were grown by dc magnetron sputtering in a vacuum system with a base pressure of 5.0×10^{-9} Torr. We deposited a series of multilayers of the following composition: thermally oxidized Si substrate/ $Ni_{80}Fe_{20}$(4 nm)/ CuNi(d_f)/ Nb(d_s = 18 nm)/ CuNi(d_f)/ $Ni_{80}Fe_{20}$(4 nm)/ $Ir_{25}Mn_{75}$(10 nm)/ Al(4 nm) with d_f ranging from 2 nm to 18 nm (see Fig. 1(a)). The ferromagnetic $Ni_{80}Fe_{20}$ layers placed next to the CuNi layers ensure uniform magnetization of the CuNi layers in our measurements [6]. The magnetization direction of the top F layer is pinned in the plane of the sample by exchange bias from the antiferromagnetic $Ir_{25}Mn_{75}$ layer. The CuNi alloy was grown by co-sputtering Cu and Ni targets, and the Ni concentration of 52 atomic percent was calculated from the Cu and Ni deposition rates.

We pattern the samples into 200 μm-wide Hall bars and perform four-point magnetoresistance measurements in a continuous flow ^{4}He cryostat. Fig. 1(b) shows resistance versus temperature for the parallel (P) and antiparallel (AP) configurations of the CuNi layers for the sample with 5 nm thick CuNi. The superconducting transition temperature difference between the P and AP configurations, ΔT_c, is 7 mK. Fig. 1(c) shows the resistance as a function of magnetic field applied along the exchange

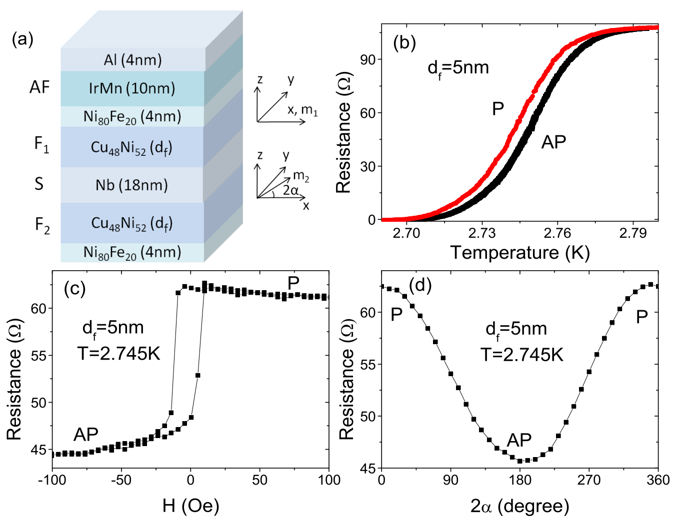

FIG. 1. (color online). (a) Schematic of the sample. Here 2α is the in-plane angle between the magnetic moments of the CuNi layers. (b)-(d) Experimental data for the sample with the CuNi layer thickness of 5 nm. (b) Resistance versus temperature for parallel (P) and antiparallel (AP) alignments of the CuNi layers. (c) Resistance versus in-plane magnetic field applied parallel to the pinned layer magnetization at $T=$ 2.745 K. (d) Resistance versus 100 Oe in-plane field angle, 2α, measured at the same temperature.

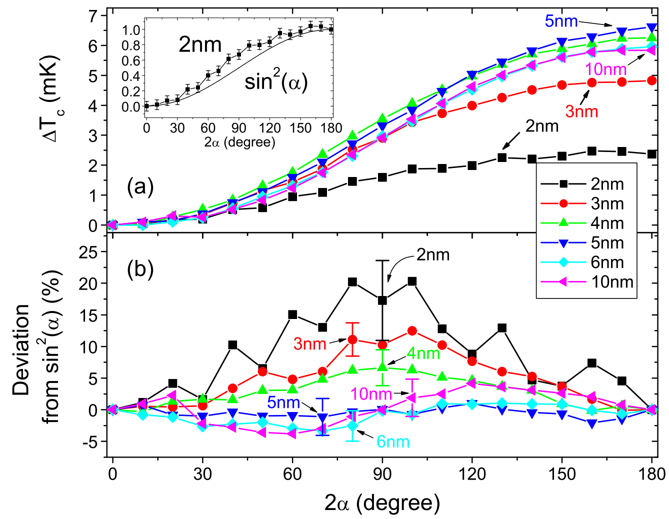

FIG. 2. (color online). (a) Angular variation of T_c defined as $\Delta T_c(d_f, 2\alpha) \equiv T_c(d_f, 2\alpha) - T_c(d_f, 0)$ for samples with the CuNi layer thickness d_f from 2 to 10 nm. Inset: normalized $\Delta T_c(d_f, 2\alpha)$ for $d_f = 2$ nm (squares) and $\sin^2(\alpha)$ (line). (b) Percent deviation of the normalized angular variation of T_c, $\Delta T_c(d_f, 2\alpha)/\Delta T_c(d_f, 180^\circ)$, from $\sin^2(\alpha)$.

bias direction in the middle of the superconducting transition at $T=$ 2.745 K. This figure demonstrates that a magnetic field of ± 100 Oe is sufficient to reverse and saturate the magnetization of the free layer and thereby create well-defined P and AP configurations. Fig. 1(d) shows resistance versus the angle between the F layer magnetizations, 2α, measured at $T=$ 2.745 K. For this measurement, we stabilize the temperature in the middle of the superconducting transition to within ± 0.1mK, rotate the 100 Oe external magnetic field in the plane of the sample and measure the sample resistance as a function of 2α.

The angular dependence of T_c is calculated from the angular dependence of the resistance, $R(2\alpha)$, such as that shown in Fig. 1(d) [23]. In order to convert the resistance data into T_c, we use the slope of the $R(T)$ curve, $(\frac{dR}{dT})$, in the middle of the superconducting transition (at $T=$ 2.745 K for the sample with 5 nm thick CuNi shown in Fig. 1(b)). The angular variation of T_c, $\Delta T_c(d_f, 2\alpha) = T_c(d_f, 2\alpha) - T_c(d_f, 0)$, is calculated for a given d_f as $\Delta T_c(d_f, 2\alpha) = (R(2\alpha) - R(0)) / (\frac{dR}{dT})$. Fig. 2(a) shows $\Delta T_c(d_f, 2\alpha)$ for samples with CuNi thickness d_f from 2 nm to 10 nm. This plot demonstrates that $\Delta T_c(d_f, 2\alpha)$ significantly deviates from a simple dependence given by $\sin^2(\alpha)$. This is quantified in Fig. 2(b), which shows the percent deviation of the normalized angular variation of T_c, $\frac{\Delta T_c(d_f, 2\alpha)}{\Delta T_c(d_f, 180^\circ)}$, from $\sin^2(\alpha)$. The deviation is significant for samples with $d_f < 4$ nm. The magnitude of the deviation reaches 20% for the $d_f=$ 2 nm sample.

We next discuss the comparison of our results with theory. The transition temperature $T_c(d_f, 2\alpha)$ can be calculated for our system, which is in the clean limit, using methods that include [15, 16] the odd triplet correlations that are generated by the presence of magnets with non-collinear magnetizations. These methods are based on self-consistent solution of the microscopic Bogoliubov-de Gennes [24] equations. The matrix elements of these equations with respect to a suitable basis containing a sufficiently large number N of elements, can be evaluated analytically as a function of d_s, α, and the other parameters in the problem. The eigenvalue problem for the resulting matrix is then numerically solved, using [16] a self consistent iterative process. To find T_c, the self-consistency equation can be linearized [25] near the transition, leading to the form $\Delta_i = \sum_q J_{iq} \Delta_q$, where the Δ_i are expansion coefficients of the position dependent pair potential [25] in the chosen basis and the J_{iq} are the appropriate matrix elements with respect to the same basis, as obtained from the linearization procedure. These matrix elements can be written as $J_{iq} \equiv (J^u_{iq} + J^v_{iq})/2$, where,

$$J^u_{iq} = \gamma \int d\epsilon_\perp \sum_n^{N_D} \left[\tanh\left(\frac{\epsilon_n{}^{u,0}}{2T}\right) \sum_m^N \frac{\mathcal{F}_{qnm}\mathcal{F}_{inm}}{\epsilon_n{}^{u,0} - \epsilon_m{}^{v,0}} \right], \tag{1}$$

$$J^v_{iq} = \gamma \int d\epsilon_\perp \sum_n^{N_D} \left[\tanh\left(\frac{\epsilon_n{}^{v,0}}{2T}\right) \sum_m^N \frac{\mathcal{F}_{qmn}\mathcal{F}_{imn}}{\epsilon_n{}^{v,0} - \epsilon_m{}^{u,0}} \right]. \tag{2}$$

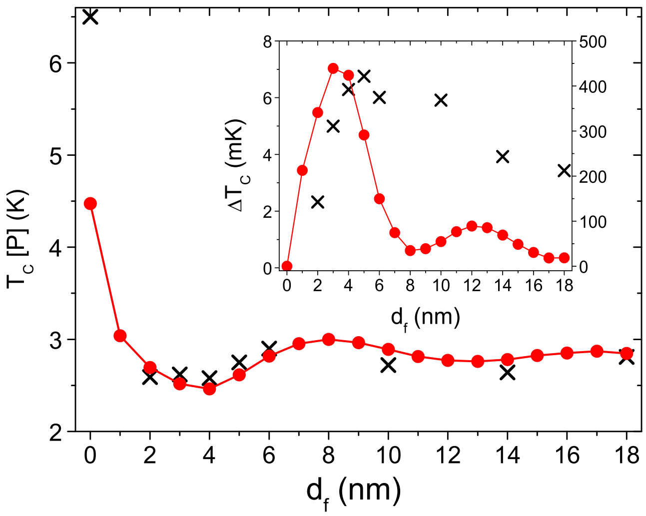

FIG. 3. (color online). Experimental data (crosses) and theoretical fittings (circles) of T_c in the P state as a function of the CuNi layer thickness d_f. The inset is the comparison of ΔT_c. Note the different scales for experimental data (left scale) and theoretical fittings (right scale).

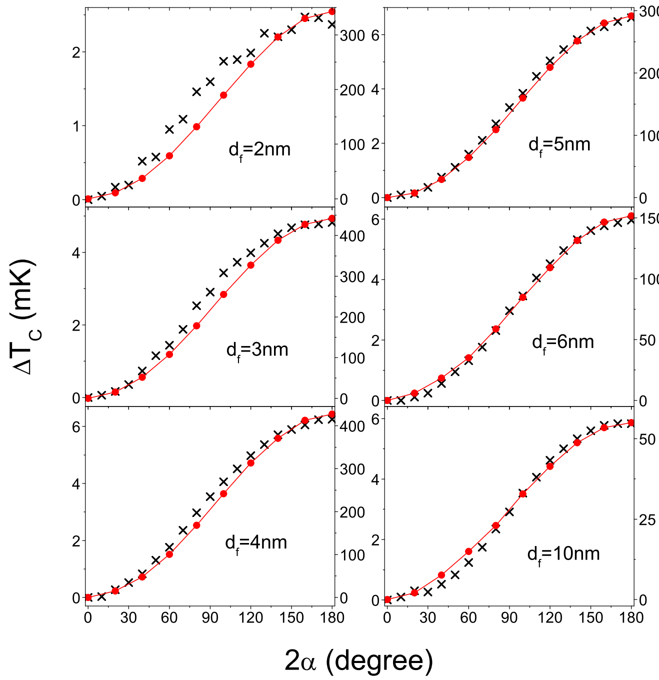

FIG. 4. (color online). Comparison between the experiment (crosses, left scale) and the theory (circles, right scale) of the angular dependence of $\Delta T_c(d_f, 2\alpha)$ for samples with different CuNi layer thicknesses d_f. For all d_f, experimental and theoretical results have similar shapes despite the mismatch in absolute values.

Here $\gamma = \gamma_0/(4\pi D)$, with γ_0 being the usual dimensionless superconducting coupling, $\epsilon_n^{u(v),0}$ are unperturbed particle (hole) energies, D is the total sample thickness d in units of the Fermi wavelength, and N_D denotes that the sum is cut off at energies beyond the Debye frequency. We have $\mathcal{F}_{qnm} \equiv \pi\sqrt{2d}\sum_{p,r}^{N}\mathcal{K}_{qpr}(d_{mp}^{\uparrow}c_{nr}^{\downarrow} + d_{mp}^{\downarrow}c_{nr}^{\uparrow})$, $\mathcal{K}_{qpr} \equiv (2/d)^{3/2}\int_0^d dz f(z)\sin(k_q z)\sin(k_p z)\sin(k_r z)$, where $k_q = q\pi/d$, the z axis is normal to the layers (see Fig. 1) and $f(z)$ is unity in the superconductor and zero elsewhere. The quantity $\epsilon_\perp$ denotes the kinetic energy due to motion in the transverse direction. The c_{ij}^{σ} and d_{ij}^{σ} are the expansion coefficients of the quasiparticle amplitudes [25] in terms of the basis set. All energies are scaled by E_F. All the sums and integrals in the above equations can be evaluated numerically with great precision as a function of temperature and of the other parameters in the problem. The value of T_c is then found [25–27] as the highest temperature for which the largest eigenvalue of the matrix J_{iq} is unity. This largest eigenvalue is readily found numerically.

In evaluating $T_c(d_f, 2\alpha)$ we have used the actual value of d_s (18 nm), and $T_{c0} = 4.5$ K, where T_{c0} is the bulk transition temperature of the material forming the superconducting layer. The only other parameters in the problem are [16, 25] the superconducting coherence length ξ_0, a parameter I characterizing the magnet strength ($I = 1$ in the half metallic limit and $I = 0$ for a non-magnetic metal) and the dimensionless barrier strength [28], H_B, characterizing interfacial scattering between the S and F layers. The intricacies of the numerics and the computing time required do not allow ξ_0, I, and H_B to be used as free parameters in a best fit procedure. We have instead explored for best values within an experimentally reasonable region of parameter values. Results presented here are for $\xi_0 = 16$ nm, reasonable for a Nb film, $I = 0.032$ appropriate for a relatively weak magnet, and $H_B = 0.75$, a barrier of moderate strength.

The results are shown in the next two figures. In the main plot of Fig. 3 we show the results for $T_c(d_f, 0)$, the transition temperature at $\alpha = 0$, versus d_f. The experimental results show the expected [25] oscillations with d_f, which are reproduced quite accurately and in detail by the theory. In the inset, we show the difference $\Delta T_c(d_f, 180^\circ)$ between the transition temperature at $2\alpha = 180^\circ$ and at $2\alpha = 0^\circ$. This quantity is also plotted as a function of d_f. Here, the general shape of the theoretical curve is qualitatively similar to the experiment, but the experimental overall scale of the phenomenon is much smaller than the theoretical one. We will return to this point below.

We consider next the dependence on 2α. In the six panels of Fig. 4 we plot experimental and theoretical results for $\Delta T_c(d_f, 2\alpha)$ as a function of 2α for six values of d_f in the experimentally relevant range. The shapes of the curves are similar to the experiment, although some deviations are present for thin ($d_f < 4$ nm) CuNi films.

However, the discrepancy in the magnitude found above at $2\alpha = 180^\circ$ affects, unavoidably, all the other angles.

Thus, the agreement between theory and experiment is in all aspects satisfactory except for the overall scale of the T_c shifts, which is much too large in the theory. This is a puzzle, since for apparently much more difficult properties such as the shape of the curves in Fig. 4 and the main plot in Fig. 3, theory and experiment are in good agreement. One would think, a priori, that while the good detailed agreement in these quantities does not preclude discrepancies elsewhere, such discrepancies could not possibly be so large. We have also attempted to fit the data to the dirty limit case, using the methods of Ref. 13. In the dirty limit, the solution is mathematically much simpler and we are able to perform a best fit to the data using six fitting parameters. Despite obtaining in some cases unreasonable parameter values, the agreement with experiment for $T_c(d_f)$ (the experimental data shown in the main plot of Fig. 3) is worse and so is the shape of the curves in the inset of Fig. 3 and in Fig. 4, particularly at smaller values of d_f. Even then, the discrepancy as to the size of the effect remains just as large and in some cases even larger than in the clean limit calculation. So, the discrepancy can not be due to the samples being closer to the dirty limit than thought. This large discrepancy in the magnitude of ΔT_c between theoretical predictions and experiment has been noted before [6, 19, 29] in other contexts.

It is natural to hypothesize that much larger surface scattering values, H_B, and smaller coherence length, ξ_0, leading to a diminished proximity effect, would drastically reduce ΔT_c. This is indeed so: we find that increasing H_B and decreasing ξ_0 sufficiently leads to values of ΔT_c of the right order of magnitude, but then the dependence of the results on d_f and α is incorrect. A possible explanation is that something in the sample interfaces weakens the normal and Andreev scattering processes responsible for the proximity effect in such a way that only the overall scale of the amplitude of the phenomenon is changed, but we cannot speculate on what this could be.

In conclusion, we made measurements of the superconducting transition temperature T_c in CuNi/Nb/CuNi trilayers as a function of the angle between the CuNi magnetic moments and the thickness of the CuNi layers. We found that T_c is a monotonically increasing function of the angle between the magnetic moments of the CuNi layers. For thin ($d_f < 4$ nm) CuNi layers, this function significantly deviates from a simple trigonometric function $\sin^2(\alpha)$. The dependence of T_c on the CuNi thickness and the functional form of the angular dependence of T_c are reasonably well described by the proximity effect theory in the clean limit but the overall scale of the angular variation of T_c given by the theory exceed the experimentally observed values by two orders of magnitude. Our measurements of the angular dependence of T_c provide new experimental constraints on theories of the proximity effect in ferromagnet/superconductor heterostructures. This work was supported by NSF Grants DMR-0748810 and ECCS-1002358. K.H. is supported in part by ONR and by a grant of HPC resources as part of the DOD HPCMP.

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[1] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Rev. Mod. Phys., **77**, 1321 (2005).

[2] A. I. Buzdin, Rev. Mod. Phys., **77**, 935 (2005).

[3] J. S. Jiang *et al.*, Phys. Rev. Lett., **74**, 314 (1995).

[4] I. A. Garifullin *et al.*, Phys. Rev. B, **66**, 020505(R) (2002).

[5] Y. Blum *et al.*, Phys. Rev. Lett., **89**, 187004 (2002).

[6] J. Y. Gu *et al.*, Phys. Rev. Lett., **89**, 267001 (2002).

[7] L. R. Tagirov, Phys. Rev. Lett., **83**, 2058 (1999).

[8] P. V. Leksin *et al.*, Appl. Phys. Lett., **97**, 102505 (2010).

[9] S. Oh, D. Youm, and M. R. Beasley, Appl. Phys. Lett., **71**, 2376 (1997).

[10] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. Lett., **86**, 4096 (2001).

[11] A. Kadigrobov, Low Temp. Phys., **27**, 760 (2001).

[12] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. B, **68**, 064513 (2003).

[13] Y. Fominov, A. Golubov, and M. Kupriyanov, JETP Lett., **77**, 510 (2003).

[14] A. F. Volkov, F. S. Bergeret, and K. B. Efetov, Phys. Rev. Lett., **90**, 117006 (2003).

[15] K. Halterman, P. H. Barsic, and O. T. Valls, Phys. Rev. Lett., **99**, 127002 (2007).

[16] K. Halterman, O. T. Valls, and P. H. Barsic, Phys. Rev. B, **77**, 174511 (2008).

[17] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Appl. Phys. A, **89**, 599 (2007).

[18] T. S. Khaire *et al.*, Phys. Rev. Lett., **104**, 137002 (2010).

[19] A. Potenza and C. H. Marrows, Phys. Rev. B, **71**, 180503 (2005).

[20] I. C. Moraru, W. P. Pratt, Jr., and N. O. Birge, Phys. Rev. Lett., **96**, 037004 (2006).

[21] J. Zhu *et al.*, Phys. Rev. Lett., **103**, 027004 (2009).

[22] G. Nowak *et al.*, Phys. Rev. B, **78**, 134520 (2008).

[23] We assume that the pinned layer magnetization is fixed and that the free layer magnetization points in the direction of the applied field. This is a valid approximation because the external field (100 Oe) is much smaller than the exchange bias field (>1 kOe) but much larger than the free layer anisotropy field (<10 Oe). We verified via quantitative modeling that this approximation does not have a significant impact on the data in Fig. [2-4].

[24] P. G. de Gennes, *Superconductivity in Metals and Alloys* (Addison-Wesley, Reading, Massachusetts, 1989).

[25] P. H. Barsic, O. T. Valls, and K. Halterman, Phys. Rev. B, **75**, 104502 (2007).

[26] K. Levin and O. T. Valls, Phys. Rev. B, **17**, 191 (1978).

[27] P. B. Allen and R. C. Dynes, Phys. Rev. B, **12**, 905 (1975).

[28] K. Halterman and O. T. Valls, Phys. Rev. B, **72**, 060514 (2005).

[29] P. Cadden-Zimansky *et al.*, Phys. Rev. B, **77**, 184501

(2008).